DESIGN AND MANUFACTURE OF HOLLOW FIBER MODULE GAS SEPARATOR

by

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ABSTRACT

Hollow fiber cellulose acetate membranes with asymmetric structure were prepared using a wet spinning method. In the preparation procedures, spinning parameter(i.e bore liquid flowrate) has been identified to influence the shape and the hollow fiber thickness. Spinneret dimension also influences the hollow fiber thickness produced. The membrane structures are characterised by using Scanning Electron Microscope. In this study hollow fiber membranes with an asymmetric structure were manufactured using a locally-designed spinneret. This paper also discusses the development of the hollow fiber membrane module.

ABSTRAK

Membran gentian geronggang selulos asetat yang mempunyai struktur asimetrik telah disediakan dengan menggunakan kaedah pemintalan basah. Dalam proses penyediaan membran ini, parameter pemintalan (kadar alir cecair rerongga) telah dikenalpasti mempengaruhi bentuk dan tebal gentian geronggang. Saiz spineret turut mempengaruhi membran gentian geronggang yang terhasil. Struktur membran dicirikan dengan menggunakan mikrograf elektron imbasan Dalam kajian ini membran gentian geronggang dengan struktur asimmetrik telah dihasilkan dengan menggunakan spinneret rekabentuk tempatan. Kertas kerja ini turut membincangkan pembangunan modul membran gentian geronggang.

INTRODUCTION

The earliest work on hollow fibres was probably begun in the very early 1960`s and came as an offshoot of work on polyester hollow fibers for textile application. This early work was aimed at gas separation via selective permeation through hollow-fibre membranes (1). At present, much of the research on hollow-fibre membranes involved asymmetric structures.

Commercial applications of hollow-fibre membranes have been established in the medical field, water reclamation (purification and desalination). Other applications are in various stages of development. A hollow-fibre is a capillary having a diameter of less than 1 mm, and whose wall functions as a semipermeable membrane (2).

Hollow fibers have become one of the most important membrane geometries, mainly because of their superior membrane area per unit of module volume and their self supporting structure. Depending on the ratio between wall thickness and outer diameter, they can withstand pressure up to 100 bar (2). In gas separation, hollow fibres are mainly used.

Hollow fiber can be prepared from almost any spinnable material. The preparation method for asymmetric hollow-fibre is closely related to that used for flat-sheet membranes (3). From a literature review it was found that the membrane preparation processes make use of phase separation and gelation phenomena, involving the demixing processes and exchange of solvent and non-solvent during the precipitation of the polymer.

Hollow fibers spun from synthetic polymers have been investigated for a number of years in the textile industry. The spinning technology developed for hollow textile fibers , particularly in the manufacture of spinnerets, has been very useful in the development of hollow fiber membranes. Hollow fiber membranes can be prepared by three conventional spinning methods (4):

(i) wet spinning (spinning from a polymer solution into a liquid coagulant).
(ii) dry spinning (spinning from a solution of a polymer in a volatile solvent into an evaporative column).
(iii) melt spinning.

In all cases, the tubular cross sections are formed by extruding the molten polymer or polymer solution through an annular of a spinneret. Spinneret design and precision of manufacture are critical features for successful hollow fiber spinning. This paper will discuss the performance of spinnerets designed by MRU from which the first hollow fiber membrane in Malaysia was produced.

SPINNERET DESIGN

A spinneret (5) is a plate containing orifices through which molten or dissolved polymer is extruded under pressure. It is either made of stainless steel, nickel alloy, platinum alloy, tantalum or glass. The orifices are often cylindrical for fibers of normal cross section.

The spinneret consist of a precision orifice containing a centrally-positioned inlet tube shown in Figure 1. The diameter of the orifice and the inlet tube are selected on the basis of the outside diameter required for the hollow fiber.

EXPERIMENTAL WORK

(1) Preparation of dope solution

The polymer that was used in this project is dry cellulose acetate(CA 398-3). Acetone (99.5%purity) and Dimethyl Formamide (DMF) is used as its solvent. Polymer solution was prepared using EASTMAN KODAKS E 400-25 formula with 26% CA,49% acetone and 25% DMF by weight. Each component with their composition is measured and mixed in a round flask using a magnetic stirrer for 24 hours until a homogeneous mixture is produced. Distillation column is used to avoid the evaporation of acetone.

2) Production of hollow-fibre membrane

Wet spinning method is used in this study as shown in Figure 2. In this process, high pressure nitrogen gas is used to transfer the polymer solution and the bore liquid through a spinneret. The flowrate of the polymer solution and the bore liquid is controlled by V1 and V4 respectively. The coagulation liquid used is deionized water with temperatures maintained between 12 - 14^{o}C . The spinneret used is a tube-in-orifice type with an outer orifice diameter of 2.5mm and its capillary diameter 1.25mm. During the spinning process, the spinneret is immersed in a coagulating bath. After the coagulation process is completed, the hollow fiber is washed and heat-annealed before it is air-dried. Several experiments were done by varying bore liquid flowrate and spinneret size. Figure 3 shows the hollow fiber produced with the spinneret used shown in Figure 4.

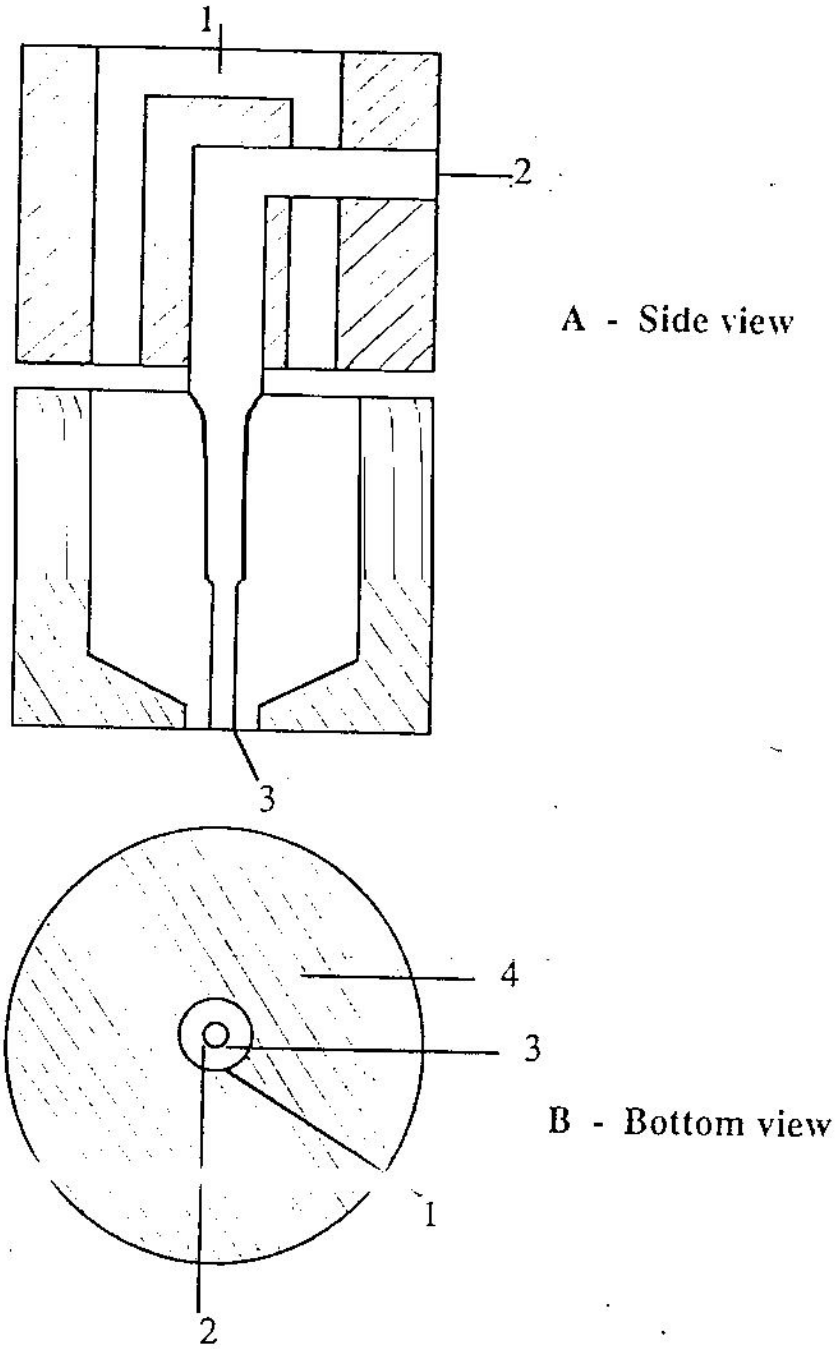
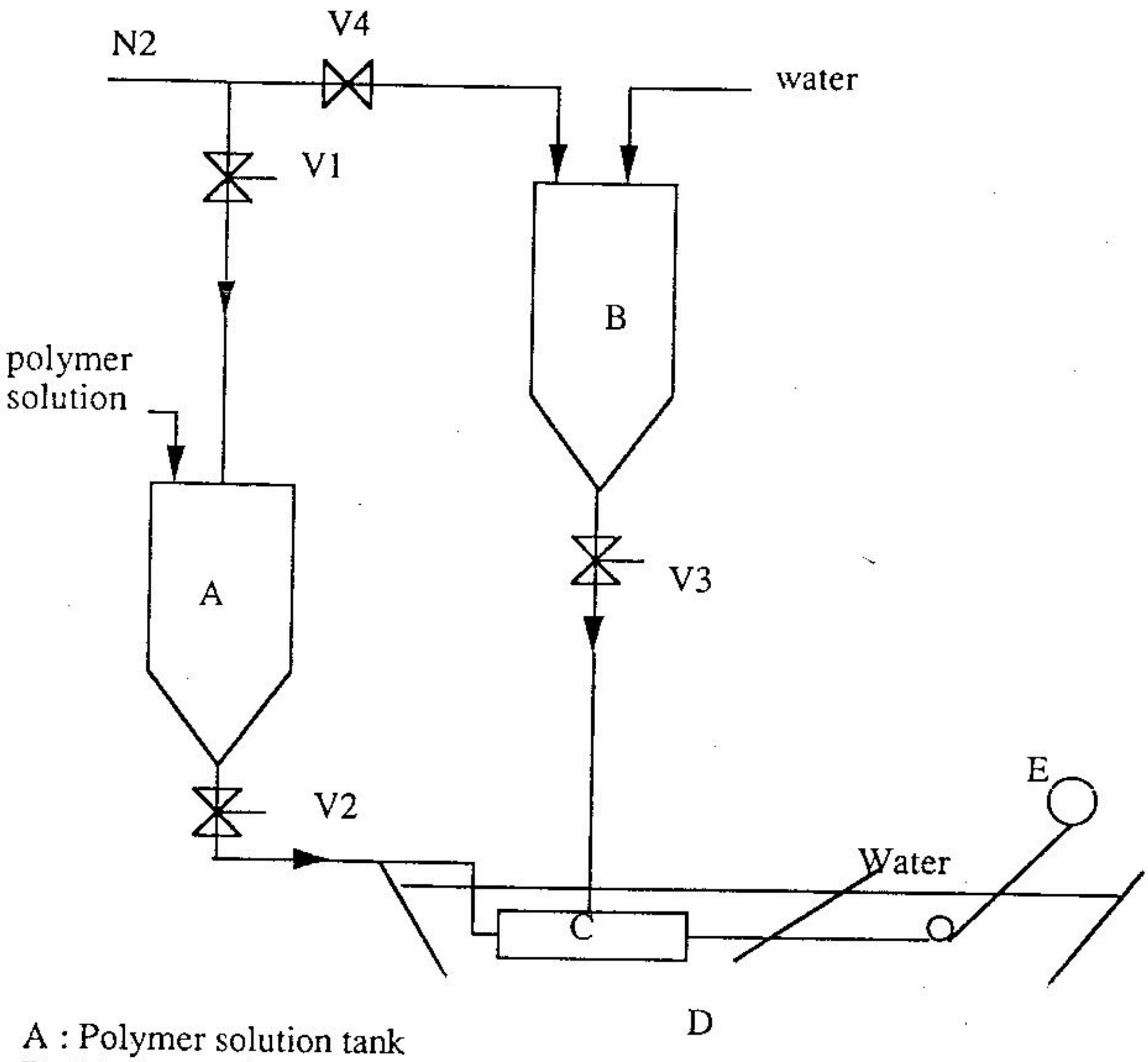


Figure 1 : Schematic diagram of tube-in-orifice spinneret.

A. Section : 1 - spinning solution entry port ;
2 - coagulating fluid entry port ; 3 - extrusion orifice
B. Bottom : 1 - spinning solution outlet ; 2 - coagulating fluid outlet
3 - inner tube ; 4 - aluminium spinneret.



A : Polymer solution tank
B : Tank containing water
C : Spinneret
D : Coagulation tray
E : Membrane rolled using roller

Figure 2 : Schematic diagram for the Production of Hollow Fiber using Wet Spinning method.

3) Determination of the membrane morphology.

The membrane morphology was examined using Scanning Electron Microscope (AMRAY 18301). Micrographs of the membrane cross-section were taken after samples were coated with a 100-300A thick gold-palladium film. Fracture samples for SEM observation of the membrane cross section were prepared as follows. A small piece of hollow fiber membrane was cut and conditioned in liquid nitrogen. The membrane was

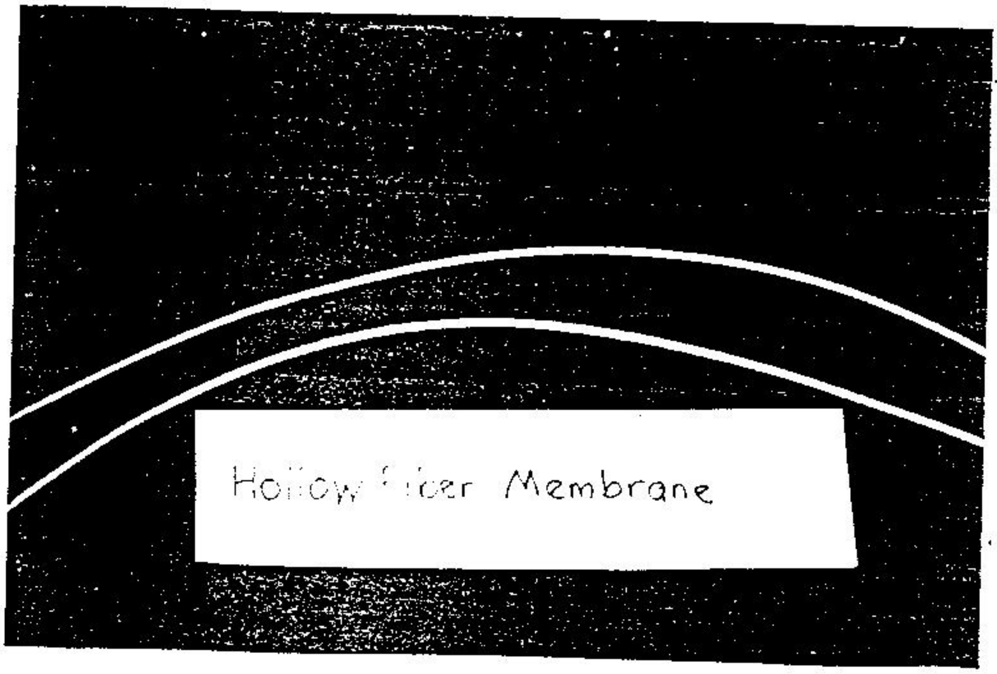


Figure 3 : Hollow fiber produce

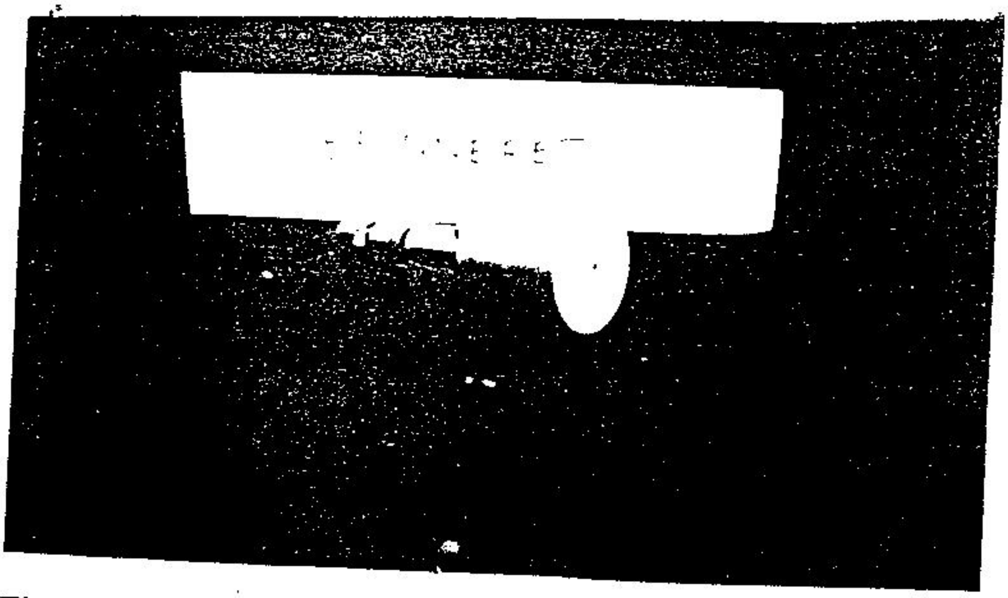


Figure 4 : Spinneret

carefully fractured at liquid nitrogen temperature. SEM pictures of the cross section of the fractured samples were taken.

RESULTS AND DISCUSSIONS

A. Morphology and Membrane Structure

Experimental data on the production of hollow fiber is presented in Table 1 and Table 2. Figure 5-Figure 11 show Scanning Electron Micrographs on the cross section of the hollow fibers. The hollow fiber produced have asymmetric structure. All the membranes produced possessed skin toplayer and a porous sublayer . Macrovoids were also formed from the external to the internal wall. Macrovoid exists because of the interactions between solvent and non-solvent and it can be observed that the interactions more pronounced indicated by bigger macrovoids near the external diameter than near the internal diameter.

B. Effect of Bore Liquid Flowrate and Spinneret Size

At constant polymer extrusion rate and take-up speed, increasing the bore liquid flowrate resulted in an increase in hole diameter and fiber outer diameter followed by a reduction in wall thickness from 743mm to 205 mm . Higher bore liquid flowrate will increase the flowrate of the polymer solution through the hollow fiber bore. This causes coagulation to occur producing a non-collapsible hollow fiber wall. Higher bore liquid flowrate also gave progressively rounder and more uniform holes. The holes is quite uniform compared to the fiber produced at lower bore liquid flowrate. The spinneret dimensions also influence the fiber diameter ; the bigger the orifice size the bigger is the hollow fiber size. In general, the hollow fiber diameter obtained is quite reasonable due to spinneret size, since the hollow fiber is spun at a very low spinning rate (i.e 0.8 m/min).

C. Module

Membrane module is the most the most critical part in membrane separation system. Separation occurs in membrane module in which the hollow fiber membranes are systematically packed to maximise the membrane area per unit volume (known as packing density). Figure 12 shows the schematic diagram of a simplest membrane modules.

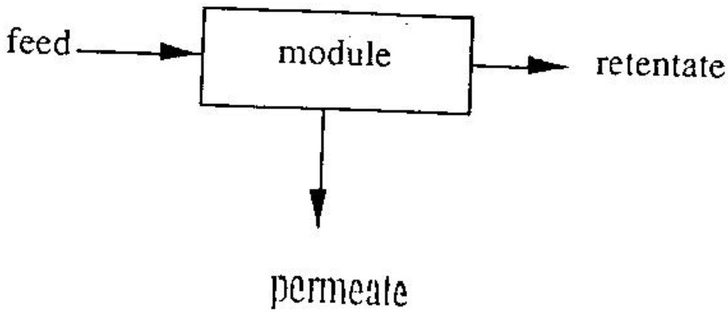


Figure 12 : A schematic diagram of a single module

Hollow fiber module is the configuration with the highest packing density, which can attain values of 30,000 m^2/m^3 (6). It is used when the feed stream is relatively clean, as in gas separation and pervaporation. Hollow fiber module has also been used in the case of seawater desalination , another relatively clean feed stream. For hollow fiber module, two types of module arrangement can be distinguished : (i) "inside-out" , where the feed solution can enter inside the fiber ; and (ii) "outside-in", where the feed solution enter the module from the outside of the fiber. A module is currently being developed in this study. Figure 13 shows the module in which the feed enters from the outside of the fiber, "outside-in". In gas separation, the module used is the "outside-in" type to avoid high pressure losses inside the fiber.

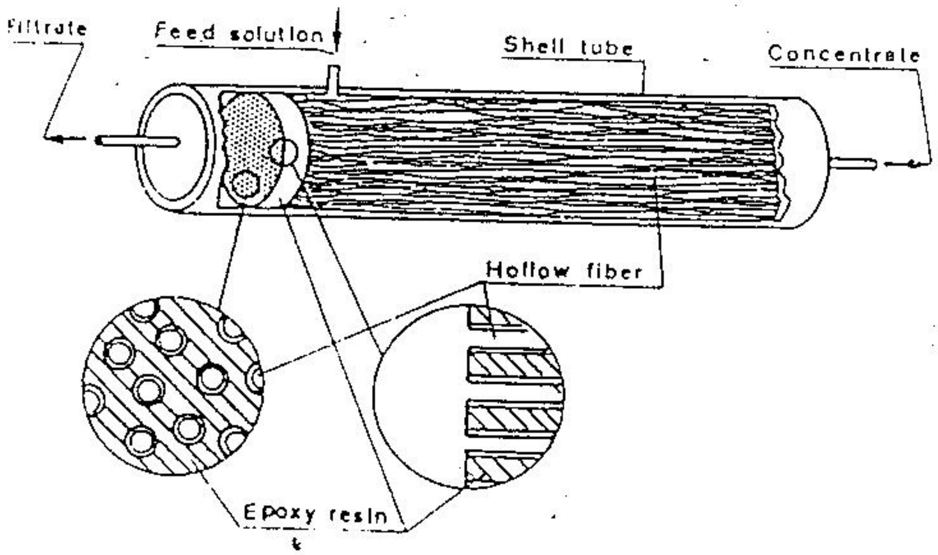


Figure 13 : Schematic diagram of a hollow fiber membrane module

Table 1 : Operating Data for the Hollow Fiber Production.

CA = 26%, Acetone = 49%, DMF = 25%.

SAMPLES	S1	S2	S3	S4	S5	S6	S7
Dope solution temp (C)	25	25	25	25	25	25	25
Polymer flowrate	40 psi(N2)	40psi(N2)	40psi(N2)	40psi(N2)	40psi(N2)	40psi(N2)	40psi(N2)
Bore liquid flowrate(cc/min)	70.0	34.28	25.7	17.0	8.5	4.0	70.0
Spinning rate (m/min)	0.8	0.8	0.8	0.8	0.8	0.8	0.8
Internal bath temp (C)	25	25	25	25	25	25	25
External bath temp (C)	12	12	12	12	12	12	12
Immersion time (min)	180	180	180	180	180	180	180
Spinnerete diameter (mm)	O.D=3.0mm I.D=1.25mm	O.D=2.5mm I.D =1.0mm	O.D=2.5mm I.D =1.0mm	O.D=2.5mm I.D =1.0mm	O.D=2.5mm I.D=1.0mm	O.D=2.5mm I.D=1.0mm	O.D=3.125mm I.D = 2.5mm
Polimer solution storage time	3 days	3 days	3 days	3 days	3 days	3 days	3 days

Table 2 : Characteristics of the Hollow Fiber produced

SAMPLES	S1	S2	S3	S4	S5	S6	S7
External fiber diameter(mm)	2.48	1.613	1.945	1.608	1.594	2.314	3.125
Internal fiber diameter(mm)	2.07	0.653	0.977	0.600	0.51	0.828	2.5
Wall thickness (mm)	205	480	484	504	542	743	313

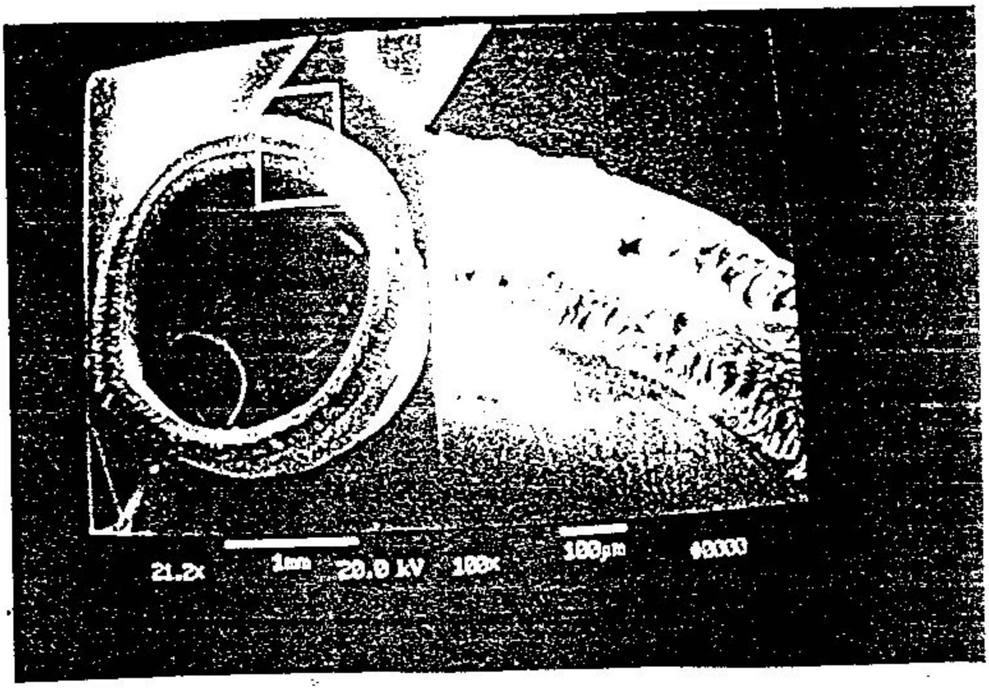


Figure 5 : Scanning electron micrographs shows the cross sections of S1

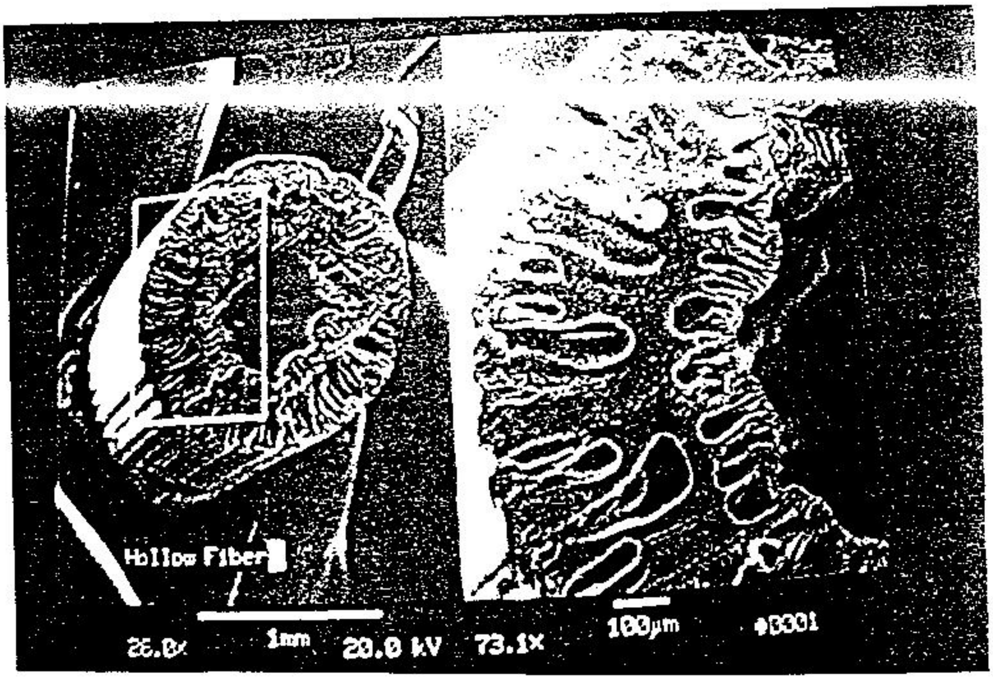


Figure 6 : Scanning electron micrographs shows the cross sections of S2

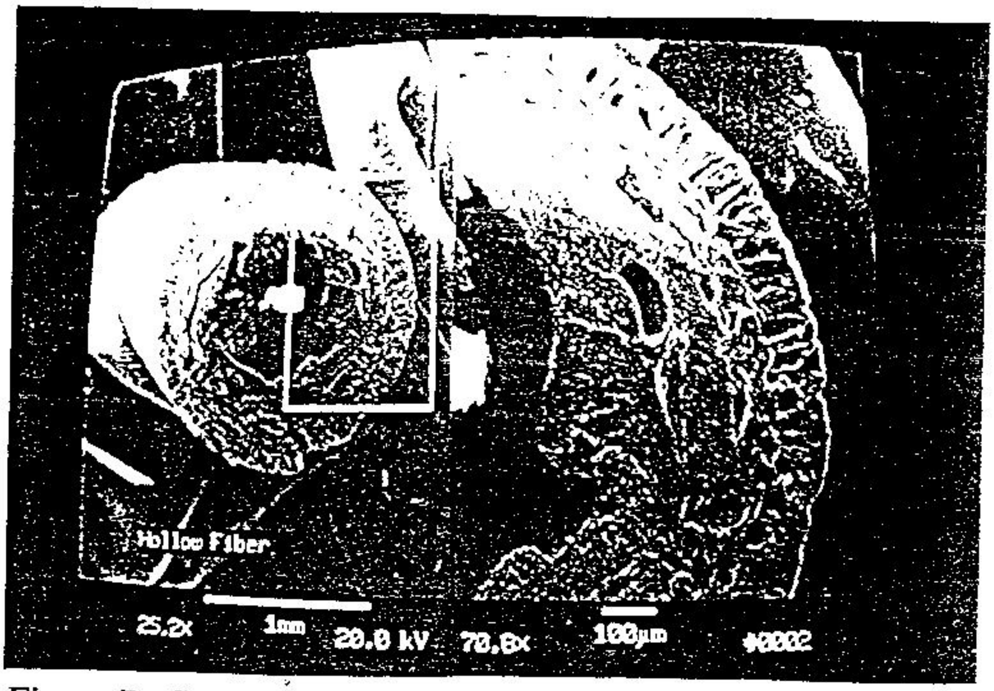


Figure 7 : Scanning electron micrographs shows the cross sections of S3

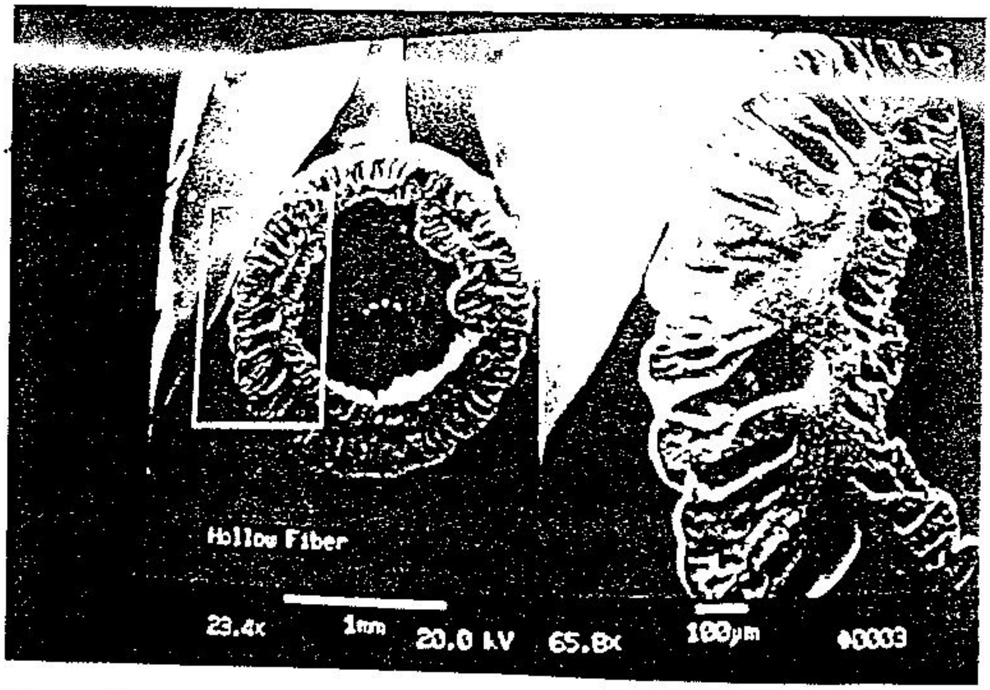


Figure 8 : Scanning electron micrographs shows the cross sections of S4

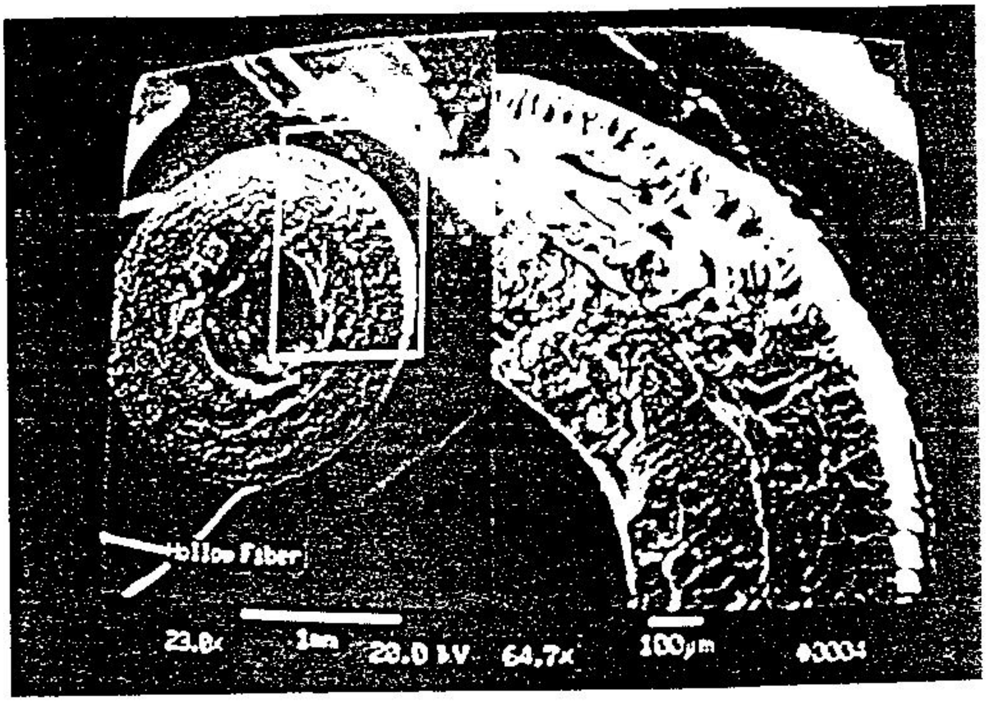


Figure 9 : Scanning electron micrographs shows the cross sections of S5

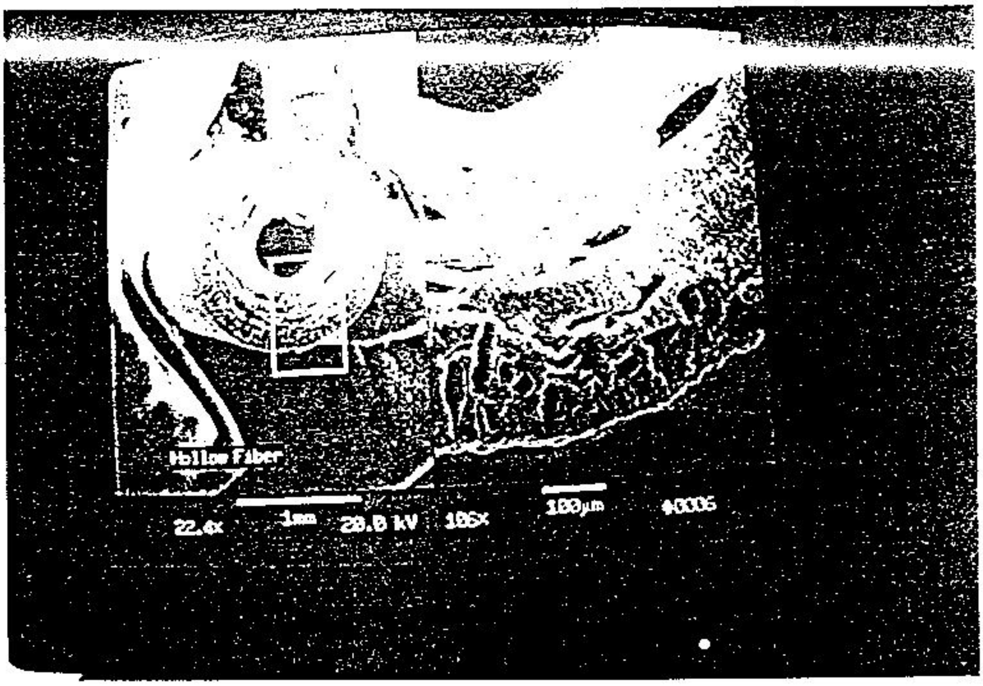


Figure 10 : Scanning electron micrographs shows the cross sections of S6

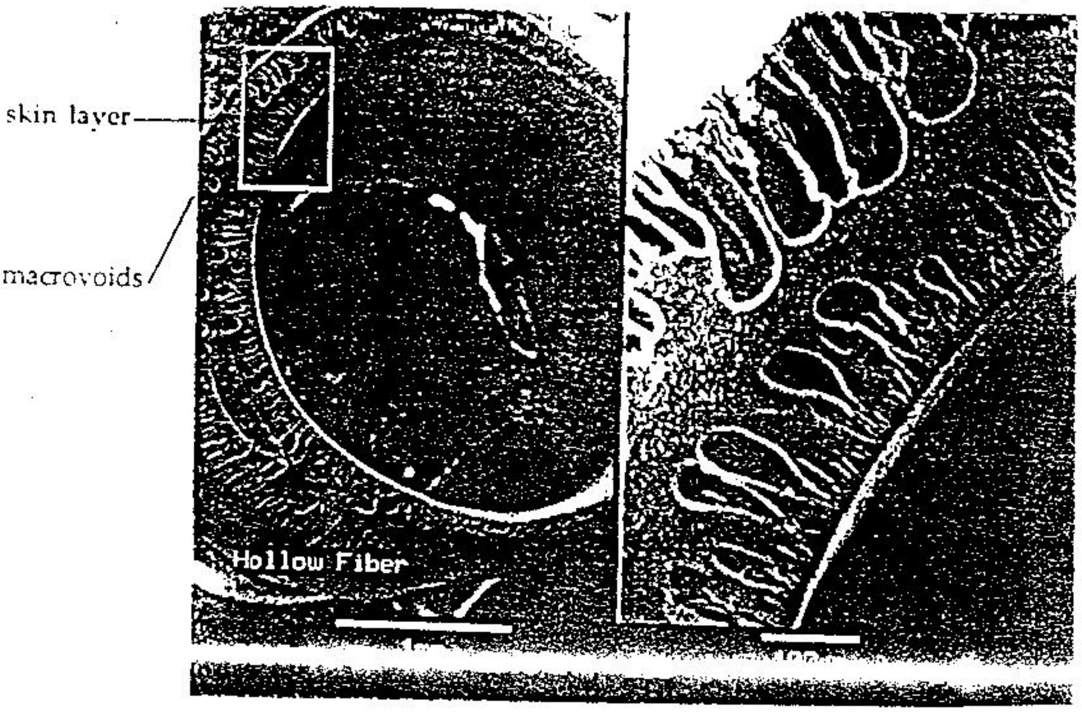


Figure 11 : Scanning electron micrographs shows the cross sections of S7

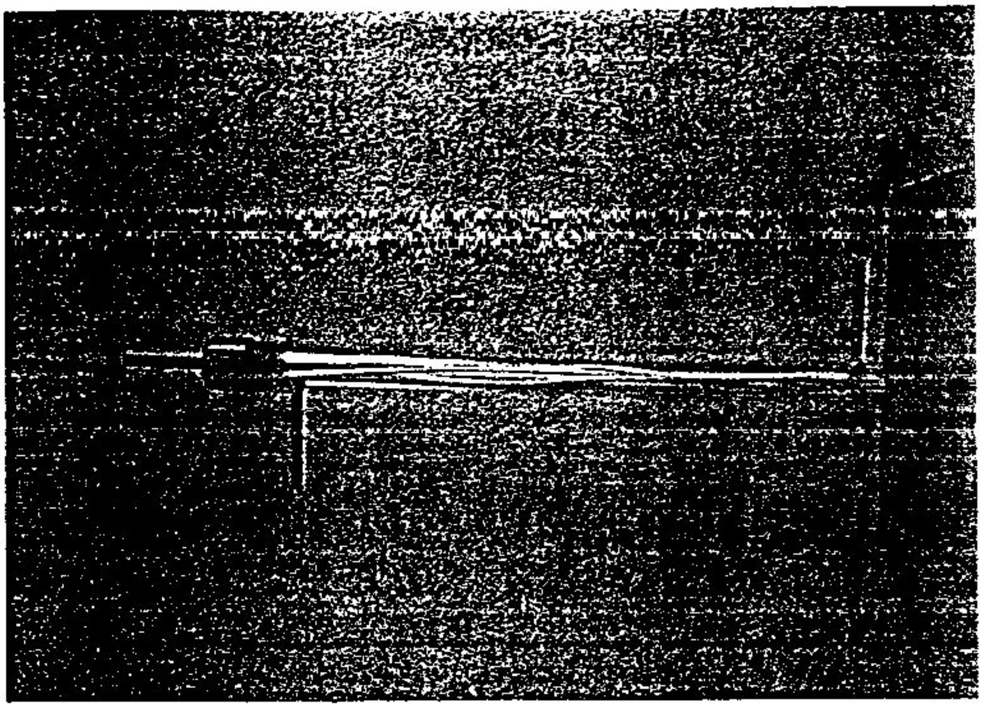


Figure 14 : Hollow Fiber Module being developed

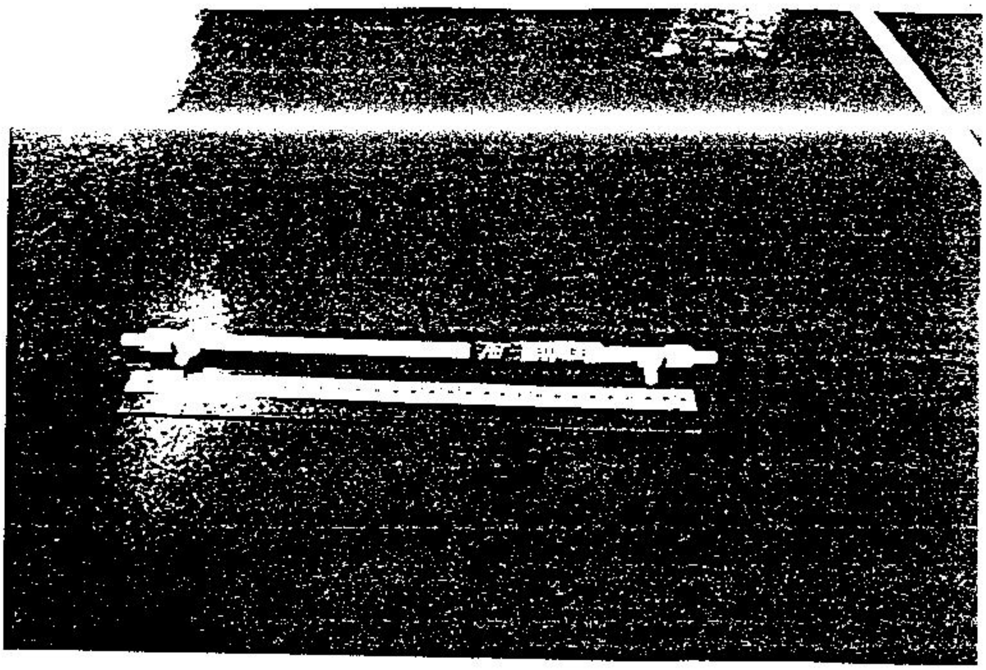


Figure 15 : Commercial Hollow Fiber Module

Figure 14 shows a hollow fiber module which is currently being developed by MRU. It is hoped that this module will perform equally or better than the readily available commercial module as shown in Figure 15.

CONCLUSIONS

In conclusion the hollow fiber produced is highly affected by its bore liquid flowrate and the spinneret size. Small diameter (<1.0 mm), hollow fiber membranes can be manufactured by using high bore liquid flowrate and small spinneret size.

RECOMMENDATIONS FOR FUTURE WORK

Based on the results and conclusions obtained from this study the following recommendations are given for future work :

1. To design a spinneret with a smaller size in order to produce a hollow fiber with a smaller diameter and wall thickness which is required for gas separation.

2. To further investigate the dependence of bore liquid flowrate and spinning rate on the diameter of the hollow fiber produced. Then a correlation can be made based on the [illegible] obtained.

3. To design a module that can withstand higher pressure for gas separation. A stainless steel tubing is suggested for this requirement.

4. To test the separation performance of the module using air or CO_2 and CH_4 mixture.

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